A novel mechanism for spin dephasing due to spin-conserving scatterings

M.W. Wu^{1,a} and C.Z. Ning²

¹ Chemistry Department, University of California, Santa Barbara, California 93106, USA

² Computational Quantum Optoelectronics, NASA Ames Research Center, M/S N229-1, Moffett Field, California 94035, USA

Received 29 June 2000

Abstract. A new spin dephasing mechanism is proposed for semiconductors with carrier momentumdependent transition energies (inhomogeneous broadening) between spin states. In the presence of this inhomogeneous broadening of the spin transitions, spin-conserving (SC) scatterings lead to irreversible spin dephasing in a complete analogy to the optical dephasing of inhomogeneously broadened optical transitions. This phenomenon is demonstrated for the case when the g-factor becomes electron-energy dependent.

PACS. 67.57.Lm Spin dynamics – 42.50.Md Optical transient phenomena: quantum beats, photon echo, free-induction decay, dephasings and revivals, optical nutation, and self-induced transparency – $78.47.+p$ Time-resolved optical spectroscopies and other ultrafast optical measurements in condensed matter

The physics and device application of the electron spin, sometimes known as spintronics, have attracted a great deal of interests experimentally [1–14] and theoretically [15–19]. Possible applications of spintronics include qubits for quantum computers, quantum memory devices, and the spin transistors etc. Understanding the mechanisms of spin dephasing is an important prerequisite for such applications.

Three mechanisms for spin dephasing have been proposed for semiconductors so far [20,21]: the Elliott-Yafet (EY) mechanism [22,23], the D'yakonov-Perel' (DP) mechanism [24], and the Bir-Aronov-Pikus (BAP) mechanism [25]. In the EY mechanism, the spin orbit interaction leads to a mixing of electron states with hole states of opposite spins. This mixing results in nonzero electron spin flip due to impurity and phonon scatterings. The DP mechanism [24] is due to the spin-orbit interaction in crystals without inversion center which results in the spin state splitting of the conduction band at $k \neq 0$. This mechanism is equivalent to an effective magnetic field acting on the spin, with its magnitude and orientation depending on **k**. Finally, the BAP mechanism [25] is originated from the mixing of the heavy hole (hh) and light hole bands due to spin-orbit coupling. The spin-flip scattering of electrons by holes via Coulomb interaction is therefore permitted, which gives rise to the spin dephasing. Very recently, Kikkawa and Awschalom found that the periodic

pump pulse train in the experiment can polarize nuclear momentum and consequently the nuclear polarization provides additional spin dephasing mechanism [13]. This dephasing mechanism is important only at low electron density and low temperature. Moreover, by sending a radio frequency signal into the sample, the nuclear polarization can be destroyed and therefore the dephasing mechanism due to nuclear polarization can be removed [26].

It is noted that all the mechanisms above either provide or are treated as spin-flip scatterings. SC scatterings, such as the ordinary Coulomb scatterings, electronphonon (EP) and electron-nonmagnetic impurity (ENI) scatterings [27] (in the following we refer them as SC Coulomb, EP and ENI scatterings) which have been extensively studied in optical problems [28], are commonly believed to be unable to cause spin dephasing alone as they commute with the total spin. The BAP and EY mechanisms, as well as the nuclear spin dephasing mechanism, provide automatically the spin-flip scatterings. The DP mechanism is treated [20,24] by extracting the anisotropic properties of **k** in different directions which, combined with the SC scatterings, give rise to the effective spin-flip scatterings.

In this paper, we propose another spin dephasing mechanism in addition to the above mentioned ones. The new mechanism results from the electronwavevector/energy dependence of the transition frequency between opposite spin states, which results in an inhomogeneous broadening of spin transitions. With

Corresponding author: e-mail: mwu@chem.ucsb.edu

this inhomogeneous broadening, SC scatterings lead to a decay of the spin coherence. This new spin dephasing mechanism is in complete analogy to the case of optical dephasing with inhomogeneous broadening in atoms [29] or in semiconductors [28].

To demonstrate the essentials of the new mechanism, we use the model proposed recently for an insulating ZnSe/Zn_{1−x}Cd_xSe quantum well under moderate magnetic field B in the Voigt configuration [17]. Based on a four spin-band model (two (spin-up and-down $\pm 1/2$) conduction bands and two $(\pm 3/2)$ heavy hole bands), we constructed the kinetic Bloch equations and calculated dephasing and relaxation kinetics of laser-pulse-excited plasma due to statically screened SC Coulomb scattering and the BAP mechanism. Differing from reference [17], here we only consider the SC Coulomb scatterings. The Bloch equations can be written as:

$$
\dot{\rho}_{\mu\nu,k,\sigma\sigma'} = \dot{\rho}_{\mu\nu,k,\sigma\sigma'}|_{\text{coh}} + \dot{\rho}_{\mu\nu,k,\sigma\sigma'}|_{\text{scatt}}.
$$
 (1)

Here $\rho_{\mu\nu,k,\sigma\sigma'}$ represents a single particle density matrix with μ and $\nu = c$ or v standing for the conduction band and heavy hole valence band respectively. The diagonal elements describe the carrier distribution functions $\rho_{\mu\mu,k,\sigma\sigma} = f_{\mu k\sigma}$ of band μ , wave vector k and spin σ . $f_{ck\sigma} \equiv f_{ek\sigma}$ represents the electron distribution function with $\sigma = \pm \frac{1}{2}$ and $f_{vk\sigma} = 1 - f_{hk\sigma}$ with $f_{hk\sigma}$ denoting the hh distribution function and $\sigma = \pm \frac{3}{2}$. The off-diagonal elements describe the inter spin-band polarization components (coherences) with $\rho_{cv,k,\sigma\sigma'} = p_{k\sigma\sigma'} = P_{k\sigma\sigma'}e^{-i\omega t}$ for the inter CB-VB polarization and $\rho_{cc,k,\frac{1}{2}-\frac{1}{2}}$ for the spin coherence. It is noticed here that for $P_{k\sigma\sigma'}$, the first spin index σ always corresponds to the spin index of the electron in the CB $(\pm 1/2)$ and the second spin index σ' to that of the hh VB $(\pm 3/2)$.

The coherent and scattering parts of the equation of motion for the electron distribution $f_{ek\sigma}$, hole distribution $f_{h k \sigma}$, optical coherence $P_{k \sigma \sigma}$ and spin coherence $\rho_{cc,k \sigma - \sigma}$ are given in detail in equations (5–13) in reference [17]. Here, for the sake of further discussion, we repeat the coherent parts of the equation of motion of electron distribution, spin coherence and optical coherence. We consider time evolution well after the pump pulse has vanished and therefore we do not include the terms relating to the pump pulse. Therefore the coherent part of the equation of motion for the electron distribution function is given by

$$
\frac{\partial f_{ek\sigma}}{\partial t}\Big|_{\text{coh}} = -g\mu_{\text{B}}B\text{Im}\rho_{cc,k,\sigma-\sigma} + 2\sum_{q} V_{q}
$$

$$
\times \text{Im}(\sum_{\sigma'} P_{k+q\sigma\sigma'}^{*} P_{k\sigma\sigma'} + \rho_{cc,k+q,-\sigma\sigma}\rho_{cc,k,\sigma-\sigma}) \qquad (2)
$$

The second term on the right hand side of equation (2) is the Fock term from the Coulomb scattering with V_q denoting the Coulomb matrix element. The coherent time evolutions of the spin coherence and optical coherence are given by

$$
\frac{\partial}{\partial t} \rho_{cc,k,\sigma-\sigma} \Big|_{\text{coh}} = \mathrm{i} \sum_{q} V_q \big[(f_{ek+q\sigma} - f_{ek+q-\sigma}) \rho_{cc,k,\sigma-\sigma} \n- \rho_{cc,k+q,\sigma-\sigma} (f_{ek\sigma} - f_{ek-\sigma}) + P_{k+q\sigma\sigma_1} P_{k-\sigma\sigma_1}^* \n- P_{k+q-\sigma\sigma_1}^* P_{k\sigma\sigma_1} \big] + \frac{\mathrm{i}}{2} g \mu_\text{B} B (f_{ek\sigma} - f_{ek-\sigma}), \quad (3)
$$

$$
\frac{\partial}{\partial t} P_{k\sigma\sigma'} \Big|_{\text{coh}} = -i\delta_{\sigma\sigma'}(k) P_{k\sigma\sigma'} - \frac{i}{2} g\mu_B B P_{k-\sigma\sigma'}
$$

$$
-i \sum_{q} V_q \left[P_{k+q,\sigma\sigma'}(1 - f_{h k \sigma'} - f_{e k \sigma}) - P_{k+q,-\sigma\sigma'} \rho_{cc,k,\sigma-\sigma} + \rho_{cc,k+q,\sigma-\sigma} P_{k,-\sigma\sigma'} \right] . \tag{4}
$$

The first term of equation (4) gives the free evolution of the polarization components with the detuning

$$
\delta_{\sigma\sigma'}(k) = \varepsilon_{ehk} - \Delta_0 - \sum_q V_q(f_{ek+q\sigma} + f_{hk+q\sigma'}) \tag{5}
$$

with $\varepsilon_{ehk} = \varepsilon_{ek} + \varepsilon_{hk}$ and $\Delta_0 = \omega - E_g$. Δ_0 is the detuning of the center frequency of the light pulses with respect to the unrenormalized band gap. The last term in equation (4) describes the excitonic correlations whereas the first term in equation (3) describes the Hartree-Fock contributions to the spin coherence.

For SC scatterings, one can show [17]

$$
\sum_{k} \left. \frac{\partial \rho_{cc,k,\sigma-\sigma}}{\partial t} \right|_{\text{scatt}} \equiv 0 \tag{6}
$$

Using equation (6) one can prove from equations $(1-3)$, that

$$
\frac{\partial^2}{\partial t^2} \text{Im} \sum_k \rho_{cc,k,\sigma-\sigma} = -g^2 \mu_B^2 B^2 \text{Im} \sum_k \rho_{cc,k,\sigma-\sigma} , (7)
$$

$$
\frac{\partial}{\partial t} \text{Re} \sum_k \rho_{cc,k,\sigma-\sigma} = 0 .
$$
 (8)

This means that the spin coherence will keep oscillating without any decay [30] in the absence of the spindephasing mechanisms mentioned above. Nevertheless, if one considers the scattering terms in equation (1) for the optical coherence, similar to equation (6), one obtains

$$
\sum_{k} \left. \frac{\partial P_{k,\sigma\sigma'}}{\partial t} \right|_{\text{scatt}} \equiv 0 \tag{9}
$$

for all the SC scatterings [17,28,31]. However, we know that these scatterings do cause optical dephasing [17,28]. The difference between the spin problem and the optical dephasing lies in the coherent part of the equations of motion. The coherent part in the equation for optical coherence, $\delta_{\sigma\sigma'}(k)$, is a function of ε_{ek} and ε_{hk} . This introduces an inhomogeneous broadening to the optical coherence. It is this broadening that causes cancellation of the phases among individual optical dipoles in the presence of scatterings that satisfy equation (9), leading to an irreversible dephasing. For spin coherence above, these is no such carrier-momentum dependent transitions in the coherent part, equation (3). Therefore, scattering events alone cannot cause any spin dephasing. This indicates that in order for SC scatterings to cause irreversible spin dephasing, inhomogeneous broadenings in spin coherence are necessary.

There are at least two ways to cause such inhomogeneous broadenings in spin coherence. The first is the DP term, as discussed in the DP mechanism in reference [18]. We will come back to this issue later in the paper. The second is through the energy dependence of the g-factor. It has been found experimentally that the g-factor is not constant. Its values at the CB edge in III-V semiconductors are smaller than the free-electron value, +2. When the electron energy is increased, the g-factor approaches the free-electron value [32]. The origin of this energy dependence comes from the nonparabolicity effect, and also the strain and the penetration effects for quantum wells [33]. The energy relaxation of electrons due to carrier-carrier scattering does not change this energy dependence of the g-factor. Within a small energy range, it was found that the g-factor can be approximated by

$$
g(E) = \beta E + g_0 \tag{10}
$$

for both 3D and 2D systems, with E being electron energy [32,34–38]. For III-V semiconductors, it was found in the experiments that β ranges from 144 to 2.2 (β and E are in units of $1/eV$ and eV respectively) whereas g_0 ranges from −51 to 1.26. It is noted that this energy dependence should not be mixed up with the many-body exchange effects of electrons [39,40]. Actually at least the lowest order of the exchange correction [39], i.e., the Fock contribution in the second term of equation (3) is totally canceled by the Hartree contribution (the first term of Eq. (3)) after the summation of k as we have proved above (Eqs. (7)) and (8)). Equation (10) introduces an energy-dependence of spin transitions, leading to inhomogeneous broadening, and thus, spin dephasing in the presence of SC scatterings.

In the following, we present the results of a model calculation to show how SC Coulomb scattering makes the spin dephasing under the inhomogeneous broadening equation (10). For the sake of comparison with our earlier results [17], we use exactly the same model as we did in reference [17]. We numerically solve the Bloch equations with only Coulomb scatterings (scatterings from the BAP mechanism are not included in this paper) to study the spin coherence of optically excited electrons in an insulating $\text{ZnSe}/\text{Zn}_{1-x}\text{Cd}_x\text{Se}$ quantum well. We apply a single circularly polarized pump pulse and calculate the time evolutions of both the optical and spin coherences together with the electron and hh distributions after that pulse under the carrier-carrier scattering. The dephasing of the spin coherence is well defined by the incoherently-summed spin coherence [17], $\rho(t) = \sum_{k} |\rho_{cc,k,\frac{1}{2}-\frac{1}{2}}(t)|$, while the optical dephasing is described by the incoherently summed polarization [28,41], $P_{\sigma\sigma}(t) = \sum_{k} |P_{k,\sigma\sigma}(t)|$. The parameters of the material and the pump pulse are exactly the same as those in reference $[17]$ except the g-factor is replaced by equation (10). To our knowledge, there is no

Fig. 1. Total densities $N_{ek\sigma}(t)$ of spin band σ for electron and N_{hk} ³/₂(*t*) for hh (solid curves) together with the incoherently summed polarization $P_{\frac{1}{2}\frac{3}{2}}$ and spin coherence $\rho(t)$ (dashed curves) are plotted against time t for $B = 4$ T. Note the scale of coherences is on the right side of the figure.

experimental and theoretical study of the value of β and g_0 for ZnSe/Zn_{1−x}Cd_xSe quantum well. It is therefore understandable that certain degree of arbitrariness exists in choosing these two numbers. In this paper we choose their values so that the final effective g -factor in the same order as measured in the experiment, to demonstrate how SC Coulomb scattering can now cause spin dephasing. It should be pointed out that the actual dephasing mechanism for this particular material may be either a combination of the effect proposed here and the BAP mechanism or the BAP mechanism alone, depending on how strong the inhomogeneous broadening equation (10) is. Moreover, it is also noticed that the linear dependence on E of the g -factor (Eq. (10)) is valid only in a small range of energy. A full investigation of the g-factor for the whole energy range and at different density difference between spin-up and -down bands is important in further understanding the spin dephasing mechanism caused by SC Coulomb scattering.

The main results of our model calculation are plotted in Figure 1. β and g_0 are taken as 97.5 eV⁻¹ and −1 respectively. The incoherently summed polarization, $P_{\frac{1}{2}\frac{3}{2}}(t)$, and the incoherently summed spin coherence, $\rho(t)$, are plotted as dashed curves in Figure 1. The total densities of each spin band $N_{e\sigma}(t) = \sum_{k} f_{ek\sigma}(t)$ for electron and $N_{h\frac{3}{2}}(t) = \sum_{k} f_{hk\frac{3}{2}}(t)$ for hh are also plotted as solid curves in the same figure. It is seen that the optical coherence is strongly dephased by the Coulomb scattering and vanishes completely within the first few picoseconds. It is further shown from the figure that the spin coherence shows strong decay. This is different from the case in Figure 3 of reference $[17]$ where g is a constant and there is no decay of the spin coherence. This verifies that the SC Coulomb scattering can cause spin dephasing

16

in the presence of the inhomogeneous broadening in the spin coherence. We also found that the dephasing is much stronger if β in equation (10) is larger.

Similar to the optical dephasing, the spin dephasing proposed in this note is a result of energy/momentumdependent transitions, which lead to inhomogeneous broadening, and thus spin dephasing in the presence of the scatterings. Differing from the BAP mechanism and the EY mechanism where the scatterings flip the electron spin and therefore $\sum_{k} \frac{\partial \rho_{cc,k,\sigma-\sigma}}{\partial t}$ $\text{BAP(EY)}\ \neq\ 0, \text{ here the}$ SC scatterings do not flip the spin of electrons (Eq. (6)). Rather, the dephasing is purely due to the phase randomization of individual spin dipoles and the resulting destructive interference.

As we mentioned before, the DP mechanism also gives the momentum dependent transitions, the Dresselhaus term in the coherent part of the spin coherence equation in reference [18]. We point out here that all the earlier treatments of DP mechanism [20,24] only considered the dephasing due to anisotropic property of the DP terms – which, combined with SC scatterings, gives rise to effective spin-flip scatterings – but overlooked the dephasing due to the inhomogeneous broadening. This can be seen from the fact that if the DP term were isotropic in all directions of k, the treatment of references $[20,24]$ would give no spin dephasing. However, one may see from above discussion that in that case one should expect spin dephasing also. A correct treatment with both the anisotropic effect and the inhomogeneous broadening effect included is presented in our recent paper for n-doped bulk GaAs [18].

In conclusion, we have proposed a new spin dephasing mechanism based on inhomogeneous broadening effect. We have shown that with inhomogeneous broadening in the spin coherence, SC Coulomb, EP and ENI scatterings can cause irreversible spin dephasing. The inhomogeneous broadening can be either the DP terms or the energy dependence of the g-factor.

MWW would like to thank Horia Metiu for his hospitality. He also would like to thank C.Z. Ning for bringing equation (9) in [31] into his attention. He is supported by NASA Ames University Consortium No. NCC-2-5368 and also partially by the National Science Foundation under Grant No. CHE 94-12410, CHE 97-09038 and CDA 96-01954, and by Silicon Graphics Inc.

References

- 1. T.C. Damen, L. Vina, J.E. Cunningham, J. Shah, L.J. Sham, Phys. Rev. Lett. **67**, 3432 (1991).
- 2. J. Wagner, H. Schneider, D. Richards, A. Fischer, K. Ploog, Phys. Rev. B **47**, 4786 (1993).
- 3. J.J. Baumberg, S.A. Crooker, D.D. Awschalom, N. Samarth, H. Luo, J.K. Furdyna, Phys. Rev. Lett. **72**, 717 (1994); Phys. Rev. B **50**, 7689 (1994).
- 4. A.P. Herberle, W.W. Rühle, K. Ploog, Phys. Rev. Lett. **72**, 3887 (1994).
- 5. C. Buss, R. Frey, C. Flytzanis, J. Cibert, Solid State Commun. **94**, 543 (1995).
- 6. S.A. Crooker, J.J. Baumberg, F. Flack, N. Samarth, D.D. Awschalom, Phys. Rev. Lett. **77**, 2814 (1996); Phys. Rev. B **56**, 7574 (1997).
- 7. C. Buss, R. Pankoke, P. Leisching, J. Cibert, R. Frey, C. Flytzanis, Phys. Rev. Lett. **78**, 4123 (1997).
- 8. J.M. Kikkawa, I.P. Smorchkova, N. Samarth, D.D. Awschalom, Science **277**, 1284 (1997).
- 9. J.M. Kikkawa, D.D. Awschalom, Nature **397**, 139 (1998).
- 10. J.M. Kikkawa, D.D. Awschalom, Phys. Rev. Lett. **80**, 4313 (1998).
- 11. H. Ohno, Science **281**, 951 (1998).
- 12. Y. Ohno, R. Terauchi, T. Adachi, F. Matsukura, H. Ohno, Phys. Rev. Lett. **83**, 4196 (1999).
- 13. J.M. Kikkawa, D.D. Awschalom, Science **287**, 473 (2000).
- 14. H. Gotoh, H. Ando, T. Sogawa, H. Kamada, T. Kagawa, H. Iwamura, J. Appl. Phys. **87**, 3394 (2000).
- 15. N. Linder, L.J. Sham, Physics E **2**, 412 (1998).
- 16. Y. Takahashi, K. Shizume, N. Masuhara, Phys. Rev. B **60**, 4856 (1999).
- 17. M.W. Wu, H. Metiu, Phys. Rev. B **61**, 2945 (2000).
- 18. M.W. Wu, C.Z. Ning, Phys. Stat. Sol. (b) **222** (2000).
- 19. S. Das Sarma, J. Fabian, X.D. Hu, I. Zutic, preprint (2000).
- 20. Optical Orientation, edited by F. Meier, B.P. Zacharchenya (North-Holland, Amsterdam, 1984).
- 21. A.G. Aronov, G.E. Pikus, A.N. Titkov, Zh. Eksp. Teor. Fiz. **84**, 1170 (1983) [Sov. Phys. JETP **57**, 680 (1983)].
- 22. Y. Yafet, Phys. Rev. **85**, 478 (1952).
- 23. R.J. Elliot, Phys. Rev. **96**, 266 (1954).
- 24. M.I. D'yakonov, V.I. Perel', Zh. Eksp. Teor. Fiz. **60**, 1954 (1971) [Sov. Phys. JETP **38**, 1053 (1971)].
- 25. G.L. Bir, A.G. Aronov, G.E. Pikus, Zh. Eksp. Teor. Fiz. **69**, 1382 (1975) [Sov. Phys. JETP **42**, 705 (1975)].
- 26. J.M. Kikkawa (private communication).
- 27. H. Haug, S.W. Koch, Quantum Theory of the Optical and Electronic Properties of Semiconductors (World Scientific, Singapore, 1993).
- 28. H. Haug, A.P. Jauho, Quantum Kinetics in Transport and Optics of Semiconductors (Springer, Berlin, 1996).
- 29. L. Allen, J.H. Eberly, Optical Resonance & two-level atoms (John Wiley & Sons, New York, 1975).
- 30. This further means that the incoherently-summed spin co- $\sum_{k} \left| \rho_{cc,k}, \frac{1}{2} - \frac{1}{2} \right| \geq \left| \sum_{k} \rho_{cc,k}, \frac{1}{2} - \frac{1}{2} \right|.$ herence defined later in the text will not decay also as
- 31. W.W. Chow, S.W. Koch, Semiconductor-Laser Fundamentals (Spinger, Berlin, 1999), p. 117.
- 32. M.J. Yang, J. Wagner, B.V. Shanabrook, J.R. Waterman, W.J. Moore, Phys. Rev. B **47**, 6807 (1993).
- 33. M.J. Yang, P.J. Lin-Chung, B.V. Shanabrook, J.R. Waterman, R.J. Wagner, W.J. Moore, Phys. Rev. B **47**, 1691 (1993).
- 34. C.R. Pidgeon, D.L. Mitchell, R.N. Brown, Phys. Rev. **154**, 737 (1967).
- 35. B.D. McCombe, R.J. Wagner, Phys. Rev. B **4**, 1285 (1971).
- 36. D. Stein, K. von Klitzing, G. Weimann, Phys. Rev. Lett. **51**, 13 (1983).
- 37. G. Lommer, F. Malcher, U. Rossler, Phys. Rev. B **32**, 6965 (1985).
- 38. H. Mayer U. Rossler, Phys. Rev. B **44**, 9048 (1991).
- 39. T. Ando, Phys. Rev. B **13**, 3468 (1976).
- 40. T. Ando, A.B. Fowler, F. Stern, Rev. Mod. Phys. **54**, 438 (1982).
- 41. T. Kuhn F. Rossi, Phys. Rev. Lett. **69**, 977 (1992).